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(54) Title: ORIENTED COEXTRUDED FILMS			
(57) Abstract The invention is an oriented multilayer film comprising at least one outer layer comprising from 50 to 100 wt.% of said outer layer of an ethylene copolymer having a density of 0.900-0.935 g/cm³ and a CDBI of 50-95 % said outer layer being in contact with a polypropylene core layer, and said film having been made by coextrusion of said ethylene copolymer and said polypropylene layer and subsequent orientation. The coextruded layers can be uniaxially oriented biaxially oriented on tenter equipment without difficulties caused by the presence of low molecular weight amorphous polymer fractions inherently present in traditional Ziegler-Natta linear low density polyethylene. The optical properties of biaxially oriented polypropylene films can be retained while the scaling temperature is lowered and the heat seal strength and the hot tack performance properties are improved.			

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ORIENTED COEXTRUDED FILMS

BACKGROUND OF THE INVENTION

5 **Field of the Invention**

This invention relates to an oriented, multilayered film comprising at least one layer of an ethylene based polymer. More specifically, this invention relates to a multilayer film made wherein at least one layer is ethylene based random copolymer, and wherein the film has been coextruded prior to being stretched for
10 orientation.

Description of the Prior Art

Oriented, multilayered films are an important class of industrial products useful principally in the wrapping or packaging of various products in commerce. In particular, oriented isotactic polypropylene film is known to be useful for its
15 high barrier properties, clarity and stiffness. Polypropylene for oriented films is typically isotactic homopolymer, or are modified polypropylene blends of isotactic polypropylene with differing isotactic polypropylene, e.g., differing melt flow rates (MFR) or tacticity, atactic polypropylene, syndiotactic polypropylene, random polypropylene copolymers having minor amounts of ethylene or higher α -olefins, and ethylene copolymers, see for example, U.S. patent 4,950,720 and WO
20 96/02386. As indicated in U.S. patent 4,921,749 and related art, other components such as petroleum resins may be added as well to modify properties such as heat sealing performance, gas permeability and stiffness. Such oriented polypropylene, particularly biaxially oriented polypropylene (BOPP), has many
25 desirable properties. However, excellent hot tack and heat seal are not among these since such films lack significant heat sealing and hot tack performance at operating temperatures suited to efficient industrial practice.

Accordingly, various polymeric layers have been used or proposed as heat sealing layers for biaxially oriented polypropylene film. Among those used
30 industrially and taught in the patent literature are polyolefins, ethylene-based and propylene-based. Preferred heat sealing layers for BOPP have in the past typically

been high propylene-content polyolefins, e.g., propylene-ethylene copolymers with 1-20 wt% ethylene, and propylene-ethylene-butene terpolymers with up to about 10 wt% each of ethylene and butene and blends of those with other olefin polymers. See for example, U.S. patents 4,643,945 (LLDPE with propylene
5 terpolymer) and 4,921,749 (propylene-ethylene copolymers) and the others listed above. These polymer selections do reduce the chemical dissimilarity of the sealing layer and core layers so as to allow good adhesion between them, but result in the requirement that subsequent heat sealing be conducted at temperatures at or above about 120 °C, requiring long contact time with heating
10 elements or high heat input to achieve the necessary temperature ranges.

Summary of the Invention

The invention is an oriented multilayer film comprising at least one outer layer comprising from 50 to 100 wt.% of said outer layer an ethylene copolymer having a density of 0.900-0.935 g/cm³ and a CDBI of 50-95% said outer layer
15 being in contact with a polypropylene core layer, and said film having been made by coextrusion of said ethylene copolymer and said polypropylene layer and subsequent orientation. Preferably the ethylene copolymer outer or skin layer, or layers, will have a MWD (M_w/M_n) of 1.8-3.5 and a MI of 0.5-10 (ASTM D 1238, 190 °C, 2.16 kg). The coextruded layers can be biaxially oriented on tenter frame
20 equipment without difficulties caused by the presence of low molecular weight amorphous polymer fractions inherently present in traditional Ziegler-Natta linear low density polyethylene. Problems of undesirable adherence to drums and jaws in processing and the presence of extractables in the films made hereby are essentially eliminated. Optical properties of biaxially oriented polypropylene films
25 (BOPP) are retained while the sealing temperature is lowered and the heat seal strength and the hot tack properties are improved. The ethylene copolymer heat seal layer thus offers a practical, economical solution for preparing improved heat seal BOPP for all of stiff, flexible and heat shrinkable packaging.

Brief Description of the Figures

Figure 1 is a graph of the heat seal and hot tack performance of the film of the examples.

Detailed Description of the Invention

5 Ethylene copolymers suitable in accordance with the invention are commercially available and can be made by essentially single-sited catalysts such as metallocene catalysts. The use of the term "essentially single-sited catalyst" refers here to any olefin polymerization catalyst that provides narrow molecular weight distribution (MWD) (M_w/M_n) and narrow composition distribution, as
10 measured by the composition distribution breadth index (CDBI). Commercially available products specifically include the metallocene LLDPE resins (m-LLDPE) marketed by Exxon Chemical Co., Houston, Texas, as EXCEED® 350D60 (ethylene-hexene copolymer, MI = 1.0, density 0.917 g/cm³) and 377L60 (ethylene-hexene copolymer, MI = 1.0, density 0.922), and the metallocene
15 plastomer ethylene copolymers marketed under the tradenames EXACT®, available from Exxon Chemical Co. and AFFINITY®, available from Dow Chemical Company, Midland, Michigan, where such plastomers have densities at or above about 0.900. Typically the ethylene copolymers will comprise as comonomers any C₃-C₁₂ α-olefin, preferably one or more C₄-C₈ α-olefin. The
20 comonomer content is established by the measured density, the density preferably being in the range from 0.910 to 0.925 g/cm³, most preferably from 0.915 to 0.925 g/cm³. Preferably the molecular weight of the invention ethylene copolymers as measured in MI (polyethylene melt index, ASTM D 1238) will be in the range from about 0.7 - 8.0, and most preferably 0.7 - 5.0. The MWD is typically about
25 2.0 - 3.5, preferably 2.0 - 2.7. The CDBI will preferably be above about 55 %, most preferably above 65 %. The melt index ratios (MIR, I_{21}/I_2) of the suitable ethylene copolymers will typically range from about 16 to about 50. Additional description of film suitable ethylene copolymers can be found in copending U.S. application Ser. No. 08/755,105, filed 22 November, 1996, the description of
30 which is incorporated by reference for U.S. patent practice.

Additional ethylene copolymer compositions suitable as sealing layers in accordance with the invention include polyethylene blend compositions wherein the ethylene copolymer defined above comprises at least 50 wt% and the complement is minor amounts of essentially compatible ethylene-based homopolymers or copolymers comprising ethylene and one or more of C₃-C₁₂ α-olefins, cyclic olefins, vinyl aromatic and polar vinyl monomers, such as norbornene, alkyl-substituted norbornenes, styrene, alkyl-substituted styrenes, vinyl acetate, acrylic acid, methyl acrylate, butylacrylate, etc. The blend compositions comprising ethylene-α-olefin copolymers are preferably those having an overall density of from about 0.910 to about 0.925 and MI of about from 0.7 to 5.0. For blends comprising the other ethylene copolymers, the copolymers are preferably those having 2-5 mol% comonomer and equivalent MI. Specific blend polyethylenes include any of LDPE, plastomers, LLDPE, MLDPE or HDPE, as those terms are used in the art, prepared by Ziegler-Natta polymerization, essentially single-sited coordination polymerization, and high pressure-free radical polymerization. Polymers comprising vinyl monomers include ethylene-vinyl (EVA), ethylene-vinyl alcohol (EVOH), ethylene acrylic acid (EAA), ethylene-methylacrylate (EMA), and the like. Blend suitable ethylene copolymers are those that have ethylene content sufficient to provide ethylene crystallinity so as to assure that the blend compositions will not be subject to gross phase separation within the blend compositions upon cooling.

Copolymers of lower crystallinity, and thus density at or below about 0.915 ("VLDPE"), including certain plastomer ethylene copolymers with C₄-C₈ α-olefins having densities from about 0.870 to about 0.915 g/cm³ are particularly suitable as blend components when present in sufficient amounts to allow increased line speeds due to the presence of the lower melting point, low density copolymers. The VLDPE copolymers will preferably be present in an amount up to about 30 wt.%, preferably from about 15-20 wt.%. Further description of the VLDPE and its uses in film applications appears in U.S. patents 5,206,075 and

5,358,792, both of which are incorporated by reference for purposes of U.S. patent practice.

Suitable polypropylene core or outer layer or layers will comprise any of the isotactic polypropylene compositions or blends known to be suitable for use as uniaxially or biaxially oriented films. The background art is instructive and is incorporated by reference for purposes of U.S. patent practice. Both of traditional Ziegler-Natta polypropylene resins and those of the newer essentially single-sited catalysts are suitable where having molecular weights sufficient to assure a match in melt viscosity suitable for coextrusion. Thus propylene polymers having melt flow rate "MFR" (ASTM D 1238, 230 °C, 2.16 kg) of from about 1.0 to about 40, preferably to 10, most preferably to 6.0 are suitable. The film grade polypropylene products ESCORENE® PP4592 E7, PP4372, and PP 4252 E1 of Exxon Chemical Co. are particularly suitable. Additionally, polypropylene random copolymers containing up to about 15 mol. % of at least one C₂ - C₈ α-olefin other than propylene will be suitable as a coextrudable polypropylene layer, as core, outer or intermediary layer.

Methods of preparing coextruded films are well known in the art. For this invention the ethylene copolymer compositions suitable as sealing layers and the polypropylene compositions are separately but concurrently extruded and joined for coextrusion prior to orientation. Because the melt viscosity of the preferred ethylene copolymers, or blends comprising them, is high enough to maintain sufficient dimensional stability at processing temperatures necessary to extrude the polypropylene film, both of thick and thin films of uniform thickness across the film surface can be maintained. The lack of low molecular weight amorphous fractions and the sufficiently high melting points of the ethylene copolymer compositions allow excellent processing without sticking to processing equipment yet while retaining the particularly suitable dimensional stability.

A preferred method of preparing the films according to the invention comprises a coextrusion process with subsequent orientation of the coextrudate. Though previously known in the industry that lower-melting point polymers can

be applied by the coating method, where the polymer is extruded in a molten or softened state onto the preformed higher softening point polymer film, it is also known that the use of chemically dissimilar polymers can result in insufficient adhesion between the layers. Thus the use of additional tie layers (or adhesive
5 layers) capable of adhering to both layers is often recommended when two different polymers are to be combined. See, e.g., J. Štepek, et al, Polymers As Materials For Packaging , Ch. 5, p. 346-349 (Ellis Horwood Ltd., 1987, English language edition).

The discovery of this invention however resulted from the observation that
10 the use of coextrusion multiple extruders in two or more streams at least one of which provides the ethylene copolymer compositions and another of which provides the polypropylene compositions, and concurrent feeding into a multi-channel die such that the two layers are in contact with each other for slit-die extrusion and subsequent orientation, can be used to prepare films that retain
15 excellent adhesion between the layers and excellent composite film properties without the need of additional tie or adhesive layers . The excellent clarity of BOPP is retained while the seal initiation temperature is decreased and the sealing properties of the film are increased both in total strength and without having to apply sufficient heating to soften the polypropylene layer. Since the sealing layer
20 polymers of the invention have softening points from about 80 to 100 °C, and melting points between about 100 and 120 °C, the effective heat sealing temperature is lower than that of typical propylene copolymer sealing layers, can be achieved more quickly in processing and results in increased line speeds in manufacturing use.

25 Films according to the invention particularly include oriented two-layer A/B films having the A sealing layer and a B outer layer of polypropylene or three-layer A/B/A films having two sealing layers A on either side of the core B polypropylene layer. Additional layers or treatments of the films in the B layer are also suitable, such known treatments as metallation or corona discharge
30 treatment with the addition of other barrier layers or tie layers are also well known

in the art to be applicable in any combination for specific articles of commerce. The A heat sealing layer can be on one side of a multilayered film, and can be used as another core layer, or on both sides to form an additional heat sealing layer. Such composite films will all benefit from the improved adherence of the A and B layers and the compatibility of the combined layered film for orientation, particularly uniaxial orientation and biannual orientation, such as by use of the tenter frame methods of orientation. Additionally, the broad plateau sealing temperatures, i.e., about 110 °C - 140 °C provide a significant advantage for industrial processes where accurate temperature controls are uneconomical are otherwise not available. The heat seals formed by pressing and heating together two such layers can fail largely by peeling within the noted plateau area when subjected to a breaking force of about 0.6-3.0 lbs. This failure mechanism provides excellent characteristics for peelable seals useful the packaging industry, particularly for flexible packaging. Higher peel strengths (>3.0 lb., e.g., 3.0-6.0 lb.) can be achieved by increasing the thickness of the heat sealing layer exemplified herein, and/or by introducing one or more additional coextruded tie or adhesive layer such as those comprising ethylene or propylene copolymers.

The polymer characterizations presented in this application were conducted under the following conditions and procedures. MI was determined as described in ASTM D 1238. All of M_w , M_n , and M_w/M_n (MWD) were determined by gel permeation chromatography (GPC) using a DRI differential refraction index detector, i.e., a Waters 150C GPC instrument with DRI detectors. CDBI was determined in accordance with the method described in columns 7 and 8 of WO 93/03093. Density was measured in accordance with ASTM D792 for both the polyethylene and polypropylene.

Examples

I. TM Long Experiments: Coextruded structures were cast on a laboratory Killion line and then biaxially oriented (6x6) on a TM Long line to prepare a 2-layer A/B film, and a 3-layer ABA film respectively comprising one and two heat sealing layers of 0.2 mil final thickness each and a polypropylene

core layer of 0.4 thickness. The ethylene copolymer heat sealing layers were extruded from EXCEED® 350D60 or 377L60 resins and the polypropylene layer was extruded from PD4252E1, all available from Exxon Chemical Co.

II. Pilot Line Experiments: The films of Tables 1-4 below were prepared on
5 a Black Clawson line and biaxially oriented on a tenter frame as described in the Tables 1-3 below. The film properties are presented in Table 4.

9
Table 1

Processing Information for Films made using Black Clawson Line*					
	A/B 10/90 (wt. ratio)				A/B/A 10/80/10 (wt. ratio)
	350D60 PD4252E1		377L60 PD4252E1		A 377L60 B PD4252E1
Film Gauge (mils)	10	20	10	20	10
A-Layer: Barrier Screw: B Extruder					
Screw Speed (RPM)	8		8		15
Output (lb./hr)	47		47		92
Melt Temp. (°F/°C)	550/		559/		560/
Load (Amps)	67		56		107
Volts	25		22		41
Head Press. (psi/kPa)	1847		1715		2950
B-Layer: Metering Screw: A Extruder					
Screw Speed (RPM)	151		141		131
Output (Pounds/hr)	423		423		368
Melt Temp. (°F/ °C)	586		585		---
Load (Amps)	128		128		123
Volts	469		431		406
Head Pressure (PSI)	4100		4266		4186
Output (Pounds/hr)					
Total	470		470		460
Line Speed (FPM)					
	50		27		43

*Screw speed and Specific output were the same for all films. 3.5 inch 30:1 L/D, 42 inch Wide Die. Air Knife Pinning. Primary Chill Roll was 87 °F (°C) and Secondary Chill Roll was 75 °F (°C) Line speed was adjusted to achieve the desired gauge. All other processing conditions were constant.

Table 2

MD Conditions used for Cast Films*			
Coextruded Films	A/B 10/90		A/B/A 10/80/10
	350D60/ PD4252E1	377L60/ PD4252E1	A 377L60/ B PD4252E1
Roll Temperature (° F)			
Preheat	270		
Slow Draw	265		
Fast Draw	265		
Annealing	250		
Cooling	68		
MD Draw			
	4/5	5	5

*Stretch Gap of 0.032 Inch. Draw Speed was Adjusted to Achieve desired MD Draw Ratio.

5

Table 3

TD Orienting Conditions used for Cast Films				
Coextruded Films	A/B 10/90			A/B/A 10/80/10
	350D60 PD4252E1		377L60 PD4252E1	A 377L60. B PD4252E1
Initial Gauge (mils)	10	20	10	20
Oven Temperatures by Oven Section				
Preheat	315°			
Stretch	315°			
Anneal	300°			
Line Speed (ft/min.)				
	60	20	60	50
TD Draw				
	6	8	6	6

Table 4

Film Properties of Biaxially Oriented Coextruded Film

Film Structure	A/B 10/90					A/B/A 10/80/10
Type Material	350D60/ PD 4252E1		377L60/ PD 4252E1			A 377L60/B PD 4252E1
Initial Gauge (mils)	10	20	10	10	20	10
MD/TD Draw Ratio*						
Engineering	4:6	5:9	3:8	4:8	5:8	5:6
Gauge Mic (mils)						
Testing Area Average	0.28	0.33	0.29	0.25	0.43	0.32
Measuretech Profiler						
Gauge Average mils	0.27	0.33	0.35	0.22	0.39	0.31
Low	0.20	0.20	0.29	0.18	0.34	0.24
High	0.39	0.54	0.52	0.28	0.58	0.43
Dart Drop Method A						
(g)	228	168	168	181	263	275
(g/mil)	814	509	579	724	612	859
Puncture (Inside) ***						
Peak Force (lb.)	15.09	17.13	14.87	12.57	22.83	14.85
Peak Force (lb./mil)	53.90	51.90	51.28	50.26	53.09	46.41
Break Energy (in-lb.)	7.60	5.17	7.07	4.72	7.43	8.54
Break Energy (in-lb./mil)	27.13	15.67	24.37	18.86	17.28	26.70
Puncture (Outside) ***						
Peak Force (lb.)	11.34	14.62	9.75	10.04	18.05	15.02
Peak Force (lb./mil)	40.51	44.31	33.62	40.15	41.97	46.94
Break Energy (in-lb.)	4.89	4.09	3.67	3.38	5.73	8.88
Break Energy (in-lb./mil)	17.48	12.40	12.64	13.50	13.33	27.75

Table 4 Continued

Film Structure	A/B 10/90					A/B/A 10/80/10
Type Material	350D60/ PD 4252E1		377L60/ PD 4252E1			A 377L60 B PD 4252E1
Initial Gauge (mils)	10	20	10	10	20	10
Tensile @ Yield (psi)						
MD	6,882	6,985	6,673	7,233	7,560	5,430
TD	15,111	42,866	24,924	27,599	38,341	11,954
At 200% MD	0	0	11,461	0	0	0
Ultimate Tensile (psi)						
MD	19,029	19,231	12,937	18,846	23,621	16,046
TD	34,750	65,936	45,231	47,845	66,173	25,988
Elongation @ Yield (%)						
MD	4.1	4.3	4.2	4.2	4.5	4.2
TD	5.9	5.6	5.3	5.6	5.9	5.8
Break Elongation (%)						
MD	133	140	239	155	142	145
TD	26	15	17	19	21	31
Initial Gauge (mils)	10	20	10	10	20	10
1% Secant (psi)						
MD	304,830	337,830	318,670	303,170	349,000	278,170
TD	461,330	1,153,670	748,670	788,170	975,330	383,330
Elmendorf Tear **						
MD (g)	1	2	2	1	2	2
MD (g/mil)	4	6	6	5	5	6
TD (g)	0	1	0	0	1	1
TD (g/mil)	0	3	0	0	2	3
Shrink (%)						
MD	57	58	58	64	72	42
TD	79	82	88	86	86	71

Table 4 Continued

Film Structure	A/B 10/90					A/B/A 10/80/10
Type Material	350D60/ PD 4252E1		377L60/ PD 4252E1			A 377L60/ B PD 4252E1
Initial Gauge (mils)	10	20	10	10	20	10
Haze (%)	0.63	0.77	1.07	0.97	0.97	1.20 *
Gloss (45°)	93	90	88	87	88	92 *
COF (kf)						
(I/I)	No Sample	.49	.46	.43	.39	> 1.0
(I/O)		.51	.43	.40	.40	
(O/O)		.52	.53	.49	.44	
WVTR (gm x mil/100" sq/day)		0.34			0.35	

* Engineering draw ratios were determined during the trial. The Actual draw ratios were determined from the films produced.

** Tear values were based on using a 200 gram pendulum. ASTM states that tear values are to be obtained from readings between 20 - 60% of pendulum weight. The tear values were so low, this was impossible.

***Used 2 slip sheets of HDPE.

III. The films of the following examples 1 - 4 were prepared by coextruding and casting a 2-layer tape, and then biaxially orienting. The casting was done on a Black-Clawson line and the orientation on a pilot tenter line. The resulting films are described in Table 5 and the conditions used to orient the films on the pilot tenter line are summarized in Table 6.

The properties of the coextruded film were similar to those of biaxially oriented polypropylene film without the heat seal layer. For example, the products retained excellent optics (haze and gloss), Elmendorf tear was essentially equivalent (.10 g/mil.). The ethylene copolymer heat seal layer was just 0.03 to 0.12 mil (0.7-3.0 μ), but microscopic examination showed that the heat seal layer drew uniformly with the polypropylene. Hot tack and sealing performance were measured along both machine direction (MD) and transverse direction (TD), see Figure 1. Though highly oriented, the seals introduced exhibited excellent appearance without observable shrinkage. For these examples the seal failure occurred at moderate force levels fully acceptable in industry, and the failure mechanism was largely by peeling rather than by tearing or breaking. The mode of failure at the exemplified moderate force levels

illustrate utility for peelable seals desired for many packaging applications. The hot tack measurement, also in Figure 1, illustrate suitable values for the heat sealing at between 110 - 120 °C, a significant advantage over those temperatures required for heat sealing layers comprising polypropylene copolymers.

5

Table 5
Examples of A/B Coextruded Structures
Biaxially Oriented on Pilot Tenter Line

Coextruded Sample #	m-LLDPE	Polypropylene	Orientation, TDxMD	Final Film Thickness, mil
1	10% 350D60	90% PD4252E1	5x9	0.33
2	10% 377L60	90% PD4252E1	5x8	0.39
3	10% 350D60	90% PD4252E1	4x6	0.27
4	10% 377L60	90% PD4252E1	3x8	0.35

10

Table 6**b. MD Orientation**

Coextruded Sample #	Eng'g. Orientation	Input Film Speed, ft/s (m/s)	Pre-heat Rolls, °C	Draw Rolls, °C	Anneal Rolls, °C	Chill Rolls, °C
1	5	10 ()	132	126	120	21
2	5	10 ()	132	126	120	21
3	4	10 ()	132	126	120	21
4	3	10 ()	132	126	120	21

b. TD Orientation

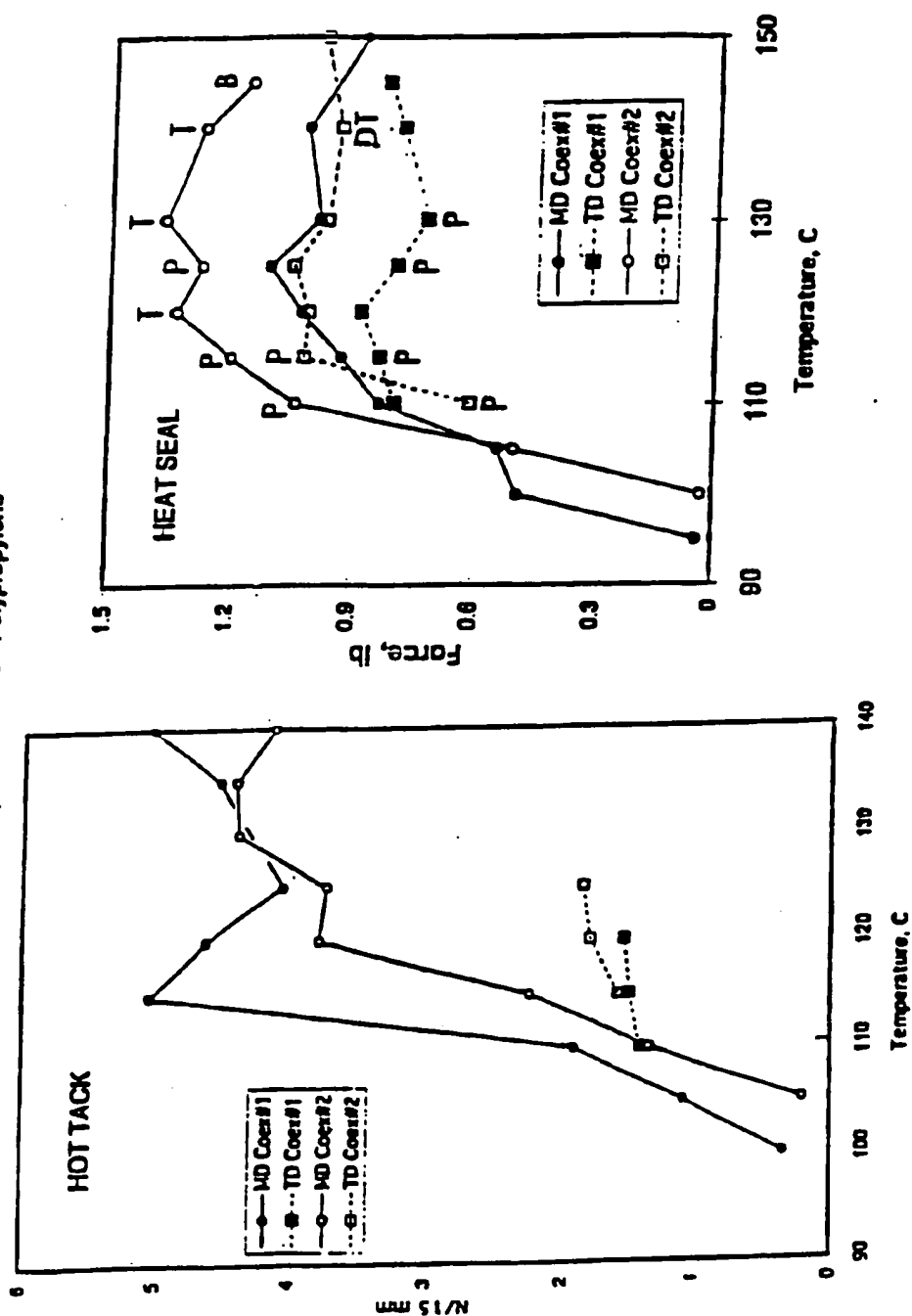
Coextruded Sample #	Eng'g. Orientation	Pre-heat, °C	Stretch, °C	Anneal, °C
1	9x	132	160	149
2	8x	132	160	149
3	6x	132	157	149
4	8x	132	157	149

Claims :

1. An oriented multilayer film comprising at least one outer layer comprising an ethylene copolymer having a density of 0.900-0.935 g/cm³ and a CDBI of 50-95%
5 constituting 50 to 100 wt.% of said outer layer, and the outer layer being in contact with a polypropylene core layer, the film having been made by coextrusion of said ethylene copolymer and said polypropylene layer and subsequent orientation.
2. The film of claim 1 wherein said film is biaxially oriented, said ethylene copolymer having a density of 0.910-0.935 g/cm³, a MWD of 1.8-3.5, and a MI of
10 0.5-10.
3. The film of claim 2 wherein said ethylene copolymer has a density of 0.915-0.925 g/cm³.
4. The film of claim 1 wherein said outer layer is a polyethylene blend composition comprising 50-99 wt% of said ethylene copolymer and 1-50 wt%
15 essentially compatible ethylene-based homopolymers or copolymers comprising ethylene and one or more of C₃-C₁₂ α -olefins, cyclic olefins, vinyl aromatic and polar vinyl monomers, said blend composition having an overall density of from about 0.910 to about 0.925 and an MI of about from 0.7 to 5.0.
5. The film of claim 4 wherein said essentially compatible ethylene-based
20 copolymer constitutes up to about 30 wt.% of the total blend and is an ethylene copolymer comprising one or more C₄-C₈ α -olefin having a density of from about 0.880 to about 0.915 g/cm³.
6. The film of claim 1 wherein said polypropylene layer consists essentially of an isotactic polypropylene homopolymer or a polypropylene random copolymer.
7. The film of claim 6 comprising two of said outer layers and at least one
25 polypropylene layer in contact with at least one of the outer layers.
8. The film of claim 1, wherein the second outer layer is polypropylene.

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A Figure 1 Hot Tack and Sealing Behavior
10% A/90% B Biaxially Oriented Coextruded Films
A=mLLDPE B=Polypropylene



INTERNATIONAL SEARCH REPORT

International Application No

PCT/US 98/03562

A. CLASSIFICATION OF SUBJECT MATTER

IPC 6 C08L23/16 B32B27/32 C08J5/18

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 6 C08L B32B C08J

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
P, X	WO 97 44178 A (UNION CAMP CORP) 27 November 1997 see page 3, line 7 - line 9 see page 6, line 13 - page 8, line 3 ---	1-8
P, X	EP 0 802 046 A (GRACE W R & CO) 22 October 1997 see page 3, line 40 - line 50 see page 8, line 18 - line 23 see page 8, line 57 - page 9, line 17; example 2 ---	1-8
X	WO 95 13321 A (EXXON CHEMICAL PATENTS INC) 18 May 1995 see page 3, line 30 - page 4, line 8 see page 5, line 26 - page 9, line 28 see page 11, line 4 - line 8 see page 11, line 20 - line 33 --- -/--	1-8

☒ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

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"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

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"&" document member of the same patent family

Date of the actual completion of the international search

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INTERNATIONAL SEARCH REPORT

International Application No

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C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	WO 92 14784 A (EXXON) 3 September 1992 see page 10, line 3 - page 11, line 19 see page 19, line 8 - line 30 -----	1-8

INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

PCT/US 98/03562

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